JOHN D. ROBERTS

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I met the *great* John D. Roberts at Caltech for the first time in 1972. He seemed larger than life: a tall, handsome, rugged man with a shock of curly grey hair, broad warm grin, huge embracing handshake, and twinkle in his eye. This was the man who has inspired a generation of organic chemists by his intellectual leadership, his innovation in developing new techniques to explore mechanistic organic chemistry, his contributions to undergraduate and graduate education and the advancement of scientific societies.

George Hammond, a brilliant pioneer in the field of molecular photochemistry and Roberts' colleague at Caltech, wrote in October, 1969:[1]

> *"For as long as my mind thinks, John Roberts will remain as a unique image therein. He is a rare man and a rarer scientist. As an academic colleague and a scientific peer, he has probably made me as angry, and yet inspired in me as much respect and admiration, as any man I have known. I first met Jack Roberts in 1945 when he arrived at Harvard as an NRC Research Fellow. Later I shared a laboratory with him for the better part of a year. Memories of that year are highlighted by three peaks: our sharing of the broadcasts of Red Sox games at a volume that had Paul Bartlett jumping out of his skin in his office across the hall; Jack's brutal evaluation of my laboratory housekeeping; and the feeling of exhilaration when Roberts engaged me in scientific argument. To debate science with Roberts is a unique experience. He combines the style of a dyspeptic porcupine and the wily cunning of a Las Vegas dealer with fundamental integrity to produce a style that is both frightening and stimulating.*
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> *"If John Roberts were only a scientific disputant, he would still have earned a place in scientific history. However, the things that he has done far outrank the things that he has said. Roberts fastened onto carbon-14 when most chemists still feared that radioactivity would sterilize them; he learned molecular orbital theory when most organic chemists regarded an operator as a bogeyman; he set a style now copied by hundreds of young men for the study of small carbocyclic compounds; he applied nuclear magnetic resonance to organic compounds when most of us were still pondering spin flipping in ice; and he emerged from a violent romance with computer science as a truly triumphant lover.*
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> *"When you read the papers by Roberts and his coworkers published in the early 1950's, you see the concatenation of a shrewd analytical intellect, the appreciation of the power of a new experimental method, and a deep perception in the choice of problems. The use of carbon-14 to probe the timing of skeletal rearrangements and protonic peregrinations in carbonium ions set a new standard for a dominant field of chemistry.*
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> *"Perhaps the most shocking single mechanistic pronouncement of our times was the elucidation of the benzyne mechanism for cata substitution in the reactions of unreactive aryl halides with powerful nucleophilic reagents. I recall one prominent American chemist who made an impassioned plea in a symposium that the benzyne mechanism be abandoned because it was an affront to both common sense and established theory of chemical bonding. Fortunately, the mechanistic model survived such attacks and has now become a potent tool in the hands of synthetic chemists.*
>
> *"Roberts' work in the field of molecular orbital calculations and nuclear magnetic resonance is of paramount importance in modern organic chemistry.*

Fig. 1 - John D. Roberts.

Today, nearly every student of organic chemistry casually feeds secular determinants to high-speed computers and admires the parameters generated by the electronic brain. Roberts was doing the job when one had to diagonalize matrices by direct use of group theory and drudgery.

"If Roberts had not entered the field of NMR at an early stage, I believe that the field would have developed differently and far less effectively. His dogged determination to do things right led him to treat the problems of spin-spin coupling and chemically induced spin relaxation in a way that was alien to the style of organic chemists. To be sure, such work would have been done, but if Roberts had not been a creative participant, organic chemists would probably have regarded complex spin-coupling problems as esoteric exercises lying within the province reserved for the physical chemical elite. To see the reality of this pacesetting activity, compare the sophisticated way organic chemists handle NMR problems with their dedicated adherence to totally empirical analysis in the fields of ultraviolet and infrared spectroscopy.

"Roberts' respect for integrity is exemplified by his professional self-consistency. Despite his wide-ranging interests, he stands like the Rock of Gibraltar as an organic chemist. Some of us wonder about the labels placed on chemists and want to redescribe our intellectual and experimental pursuits. However, we recognize that the field of organic chemistry must last at least as long as John Roberts, since he will continue to define some of the boundaries of the field by his own activities."

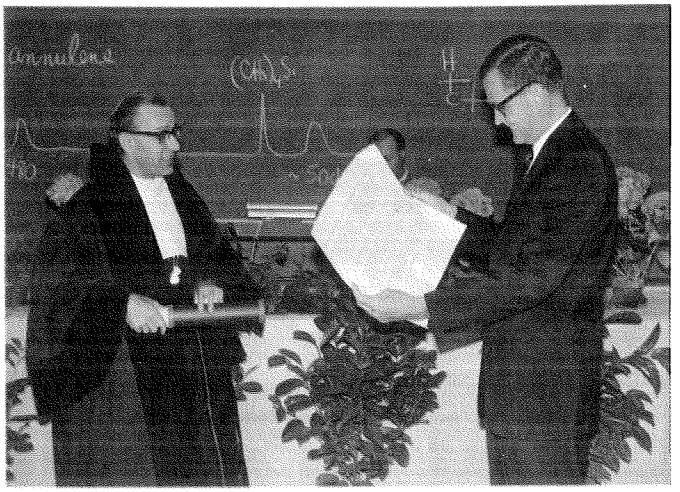

Fig. 2 - Roberts received an honorary degree at the Universität München (1962).

John D. Roberts was born in Los Angeles on June 8, 1918. From his father, Allen, he inherited a mechanical bent and great curiosity; from his mother, Flora, a love of music and literature. At the age of 10 he suffered a hearing loss. As a young boy he was an avid book reader, especially of the lives of scientists. To this day, Roberts writes with a beautiful story-telling style. Jack went to college at UCLA shortly after the Great Depression and all expenses for college were earned by working nights and Saturdays at a bakery store.[2] In 1942 he married the beautiful Edith Mary Johnson and they would raise four wonderful children: Anne, Donald, John Paul and Allen.

Fig. 3 - As a young lad, Roberts showed a keen interest in electronics.

Roberts had the benefit of many inspirational scientific mentors at UCLA and Caltech that undoubtedly shaped his early career toward organic chemistry. In the 1930's, W.R. Cromwell at UCLA gave undergraduate Roberts his first shot at research as a sophomore. Roberts went on to publish his first paper jointly with Cromwell and Caltech's D.M. Yost.[3] Organic chemistry at the time was greatly influenced by Caltech's Howard Lucas, the Ph.D. mentor of UCLA Professor William Young who would become Roberts' Ph.D. advisor, and UCLA Professor Saul Winstein, one of the great intellects of that era. Roberts' first mentor was Professor Frank Whitmore, a talented physical organic chemist at Penn State. Whitmore was extraordinarily well-organized and required monthly research reports from all his students.[2] Although Roberts only stayed at Penn State 4 months, due to the attack on Pearl Harbor, the "*monthly research report*" later became a Roberts tradition in training his own research students at MIT and Caltech.

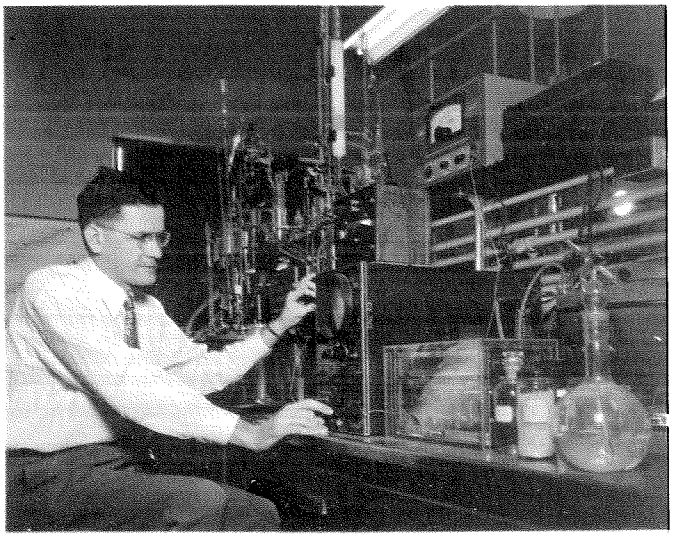

Fig. 4 - Roberts' early research experience at UCLA shaped his career toward organic chemistry.

Upon returning to UCLA to work on the war effort, Roberts wrote his thesis in 11 months and took no formal courses! He must have decided that this was not so bad. At Caltech, Roberts had been a strong proponent of getting graduate students (and undergraduates) out of the classroom and into the research laboratory as early as possible.

During his Priestley Medal address in April, 1987, Roberts tells us with characteristic modesty how lucky he was, as he puts it, *"to be in the right place at the right time."*[4]

"Never being much good at physics or mathematics, I feel I was very fortunate to get started in chemistry during a period when it was a much more descriptive science than it is today — when a knowledge of glass-blowing was more important than a knowledge of electronics or quantum mechanics; when slide rules and log tables were our computers; and when the fanciest instrument in the organic laboratory was a refractometer or possibly a polarimeter. Those things I could understand.

"Of course, a lot of other people were also fortunate in starting in chemistry in 'the good old days', but I was additionally fortunate in starting my undergraduate work at UCLA in 1936. At that time, UCLA had no Ph.D. program, but it was on the verge of getting one. As a result, UCLA was able to hire bright, young chemistry faculty, eager to do research. And those faculty encouraged me to get into research early — in fact, at the end of my sophomore year. This was very important to me, because, although I was no great shakes at course work, it turned out that I was pretty good at research and I loved it. Almost for the first time in my life, I did something really well.

"With only a few M.S. candidates and a growing undergraduate enrollment, UCLA was also short on teaching assistants. And again I was fortunate, because I was allowed to be the equivalent of a graduate teaching assistant — all told, in six different undergraduate courses. I was not the only one to profit from this particular golden period at UCLA. During that time, the school produced seven future members of the National Academy of Sciences and of those, two became Nobel Prize winners in chemistry, and two became presidents of the ACS. Not bad.

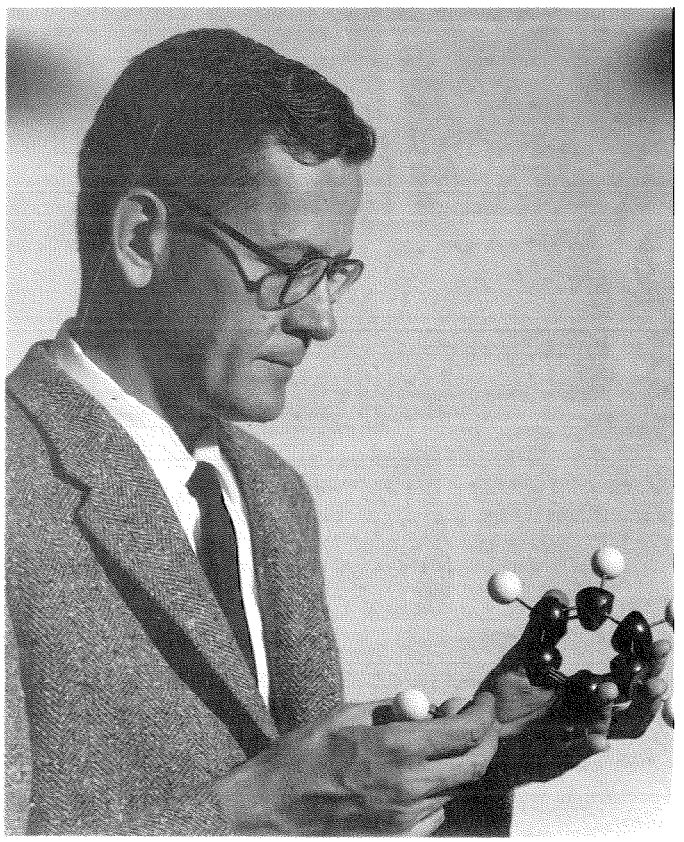

Fig. 5 - Compelling evidence supporting the intermediacy of free benzyne came from Roberts' ^{14}C isotope-labeling experiments.

"Although I finished UCLA with four rather decent undergraduate research publications, these were not enough to overcome a spotty scholastic record and get me admitted to Wisconsin for graduate study. However, Penn State was willing to take a chance and I was again fortunate, even if for only a brief period because of the start of World War II, to work with Frank C. Whitmore — a remarkable organic chemist, who became a lifelong inspiration.

"At the end of my war research, UCLA had gotten their Ph.D. program going and I was fortunate again to have a really bangup Ph.D. project with William G. Young, who himself later became a Priestley Medalist. The frosting on the cake of my graduate period was to be able to interact in a very close way with Saul Winstein, a physical organic chemist of remarkable scholarship, imagination and intellectual tenacity.

*"Then I was indeed lucky, **not** to be offered a job at du Pont, but to instead go off to Harvard as a postdoctoral fellow, just at the time R.B. Woodward was getting started there and when Paul Bartlett and Louis Fieser were in their prime. It was a confidence builder to find out that a country boy from the far West could more or less hold his own among the Harvard graduate students and postdoctoral fellows. Bartlett and Woodward helped greatly to shape my perception of what one's objectives should be in research, and the Harvard year was a great experience. It was easy to appreciate how lucky I was to be there.*

"And yet, I certainly can't claim to have always recognized good fortune immediately when it came my way. Thus, I had hoped, after my Harvard year, to get a teaching position at Berkeley and was disappointed, even a bit dismayed, when the only opening turned out to be at MIT, where Arthur C. Cope and colleagues like John Sheehan and Gardner Swain turned out to be good fortune beyond belief. And I was, and I am still, very grateful to MIT for the opportunity I was given there to get a research program underway. Admittedly, it was a bit ungracious to leave in 1953, but I felt I repaid MIT in spades by persuading Art Cope to sign up George Whitesides for a faculty position almost a year before he got his Ph.D. at Caltech. Still, it was painful to leave Cambridge just as another of my heroes, Frank Westheimer, was moving from Chicago to Harvard. But the culmination of my academic good fortune was to be offered a professorship at Caltech — a small institution, but one with a lot of clout. When I travel and meet people, they often ask how large is the Caltech student body. I always ask back — 'How large do you think it is?' The answer usually ranges from 10,000 to 40,000 and the

Fig. 6 - An underpinning to Roberts' success in proton, ^{13}C and ^{15}N NMR spectroscopy was the outstanding research spectrometers built by Varian and Bruker.

truth of about 1500 comes as a shock. Caltech turned out to be the ideal place for me to do science.

"Of course, I have had my share of missed opportunities. Somewhere around 1951, Richard Ogg of Stanford tried to convince me that nuclear magnetic resonance (NMR) spectroscopy was going to revolutionize chemistry. Being nearly illiterate in electricity and magnetism, I did not even understand what he was talking about. Four years later, I was fortunate, in the course of my du Pont consulting, to have William D. Phillips show me what NMR could do when applied to specific organic structural and rate problems. Only then did I realize how right Richard Ogg was. At that point, I didn't care whether I would ever know how NMR worked, I just knew it would solve problems that I was interested in, and, with the help of Linus Pauling, the Caltech administration, bless them, came up with the funds to buy the first commercial NMR installation in a university. And so I was able to ride the early crest of the NMR wave which has swept along through chemistry and biochemistry, as well as into medicine, with growing intensity and importance for almost forty years.

"I was also fortunate to be able to ride the early wave of the application of molecular orbital theory to organic chemistry. The later molecular-orbital waves have gotten so big, so steep and so hard to ride that I've been happy to stay on shore. Still around 1950, Hückel molecular-orbital theory (the simplest kind) had been cleverly, and carefully, kept secret by the theorists from organic chemists. 'Too tough mathematically for you guys' was the watchword. But, one day, I was lucky to look over the shoulder of my friend, William G. McMillan, one of the high priests of theory, and find to my surprise that he was using **simple algebra** *to solve a molecular-orbital problem I was interested in. I said, 'Hey, what's going on here! I can do that too!' Talk about being at the right place at the right time! So wisdom not only comes with good fortune. Sometimes you need good fortune to hit you over the head!"*

A central issue in the field of organic chemistry in the 1940's (as it is today) was understanding reactivity in organic molecules. What do we mean when we write chemical structures on paper? The scholars of the day in the growing field of physical organic chemistry were trying to come to grips with this problem. Part of the complex story unfolded in the area of carbocation rearrangements. At MIT, Roberts discovered that the rate of solvolysis of cyclopropylmethyl chloride is *exceptionally* large for a primary halide.[5] Moreover, cyclobutyl chloride is more reactive than expected. What is so special about the $C_4H_7^+$ cation! Roberts recognized early on that science is technique driven. He brilliantly invented a ^{14}C *"isotope-position rearrangement"* experiment which allowed him to conclude that the *bicyclobutonium* cation is insufficient to explain the data.[6] After late-night discussions with Woodward,[2] Roberts postulated the more symmetrical *tricyclobutonium* cation and dubbed this a *"nonclassical"* cation[6] — a term that would be much used, abused, and debated over the next few decades. Much of the work Roberts did on the $C_4H_7^+$ cation placed him as a

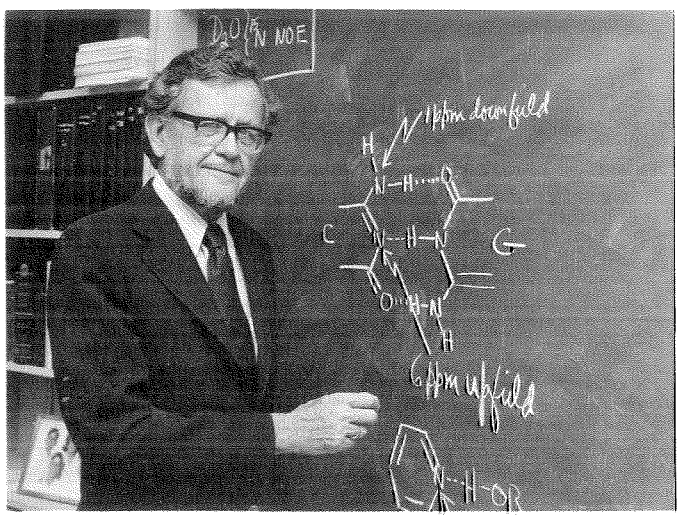

Fig. 7 - The extraordinary range of ^{15}N chemical shifts and their sensitivity to the state of protonation provides a powerful method for mechanistic studies on biologically interesting high-molecular-weight molecules such as proteins and nucleic acids.

central figure (referee?) in the public debate over nonclassical carbocations between Saul Winstein and Herb Brown. Later, George Olah (now at USC) would stun the world with proton- and ^{13}C-NMR spectra of concentrated solutions of carbocations. Olah's spectacular work ruled out the symmetrical pyramidal structure of the $C_4H_7^+$ cation (tricyclobutonium) and pointed more likely to a set of rapidly equilibrating bicyclobutonium cations. The ^{14}C isotope-position rearrangement method would again be brilliantly

applied by Roberts to reveal the intermediacy of benzyne as a free species in solution.[7]

Although to some Roberts seems like an intimidating figure (he is really a teddy bear), he is a man who rejects obfuscation as a matter of integrity. It does not surprise me that the same man who professes a weak background in physics and mathematics had great success in helping to popularize for organic chemists, molecular orbital theory and nuclear magnetic resonance spectroscopy, which were the province of the physical chemistry community in the 1950's. Roberts' books on both molecular orbital theory[8] and NMR[9,10] were outstanding pedagogical examples of demystification that opened the doors for organic chemists.

I have watched Roberts in faculty meetings. Nothing gets by him with regard to following the rules of the faculty handbook. A superficial analysis might lead one to consider that Roberts is conservative. Although this may be true, he is an adventuresome, forward-looking person as evidenced by his forays into ^{14}C labeling, NMR spectroscopy and molecular orbital theory in the 1950's. Roberts brought the first woman Ph.D. student, Dorothy Semenow, to Caltech, a move which was overdue.

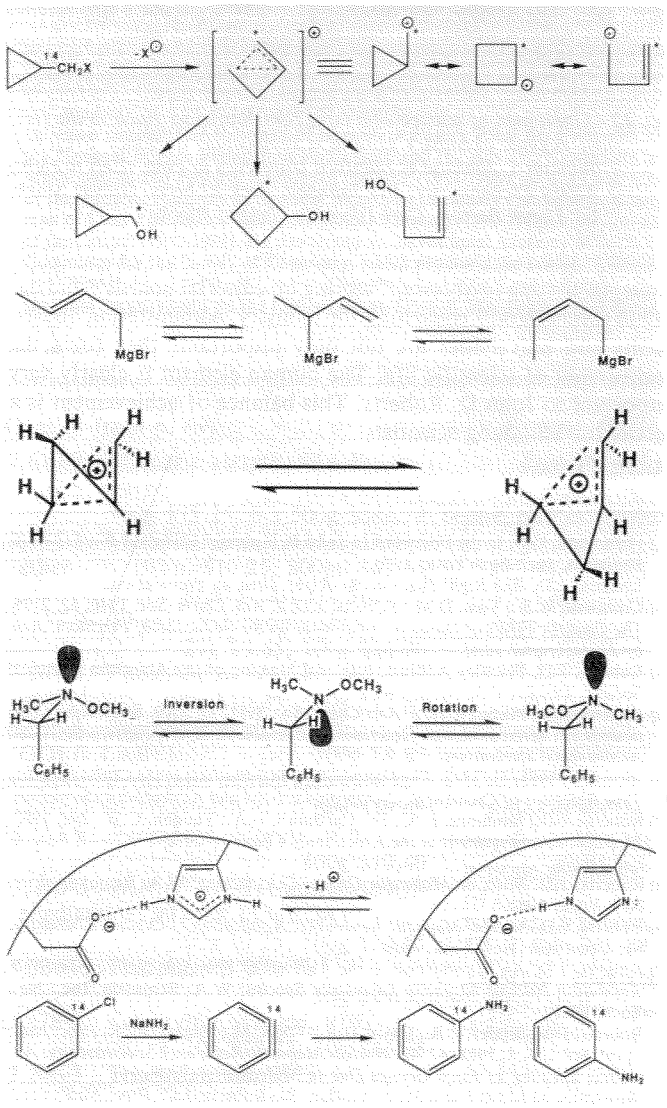

Fig. 8 - Roberts' pioneering research (isotope-position rearrangements, nonclassical cations, conformational analysis by NMR, mechanism of serine proteases, and benzyne) has merged with the body of knowledge in organic chemistry.

When Linus Pauling brought Roberts to Caltech in 1955, he convinced the Board of Trustees to buy Jack one of the first Varian NMR spectrometers. Exciting discoveries were made in those days, such as the magnetic nonequivalence arising from internal molecular asymmetry, and it became apparent that this new spectroscopic tool would allow *direct measurement of rates of rotation* about *single* bonds in ethane derivatives.[11,12] Roberts was a big hit at the 1958 Reaction Mechanisms Conference when he lectured to the physical organic community about the potential that lay ahead in this field. After intense productive research using proton NMR to study the equilibrium distribution of rapidly interconverting butenyl Grignard reagents,[13] and a conformational analysis of the structure of simple cyclic hydrocarbons,[14] Roberts convinced Varian to build a spectrometer for natural abundance ^{13}C. The potential of this became evident with the important publication from the Caltech group reporting 25 distinct resonances of 27 carbons of cholesterol.[15] Despite the fact that natural abundance ^{15}N is about a millionth of the signal strength of protons, Roberts recognized the extraordinary range of ^{15}N chemical shifts (900 ppm) 100 times that of the ordinary shift range of protons.[16] During the 1970's, Roberts demonstrated with his Bruker WH-180 the powerful role that ^{15}N spectra could have with biologically interesting high-molecular-weight molecules such as proteins and nucleic acids. This culminated in an incisive mechanistic inquiry in the "charge relay" mechanism postulated for the hydrolysis of peptide bonds by serine protease.[17]

After service to Caltech as Vice President, Provost, and Dean of the Faculty in 1980-83, Roberts has been studying the applications of NMR to medicine (magnetic resonance imaging) with a world-class team in Pasadena's Huntington Medical Research Institute.

Despite Roberts' extraordinary committment to science, he has always maintained a great interest in people and the world around him, especially the great outdoors. At Caltech he is a dynamic figure on the tennis courts, frequently seen smashing a vigorous forehand at Harry Gray or Rudy Marcus in the cool midsummer mornings. For many years he was part owner of a boat (yacht?) with Don Cram and sailing to Catalina Island was grand fun. Summers have been occupied with treacherous white-water rafting in Colorado and Chile with Edith and his sons. Winter holidays are downhill

Fig. 9 - Jack and Edith.

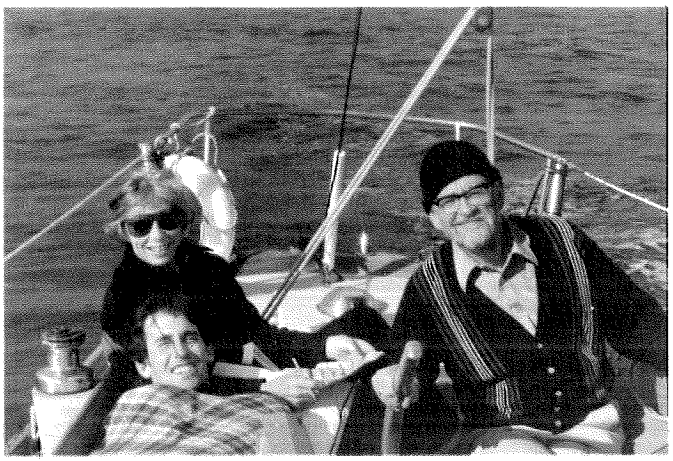

Fig. 10 - Roberts' family sailing off Catalina Island.

skiing and science discussions at Nelson Leonard's hideaway in Snowmass Village, Colorado. On a typical day at Caltech's Athenaeum, Roberts can be seen at a round lunch table for eight embroiled with engineers, physicists, geologists and biologists, in intense discussion about the design of future sailing vessels for defense of the America's Cup or his most recent lecture trips to China or safari in Kenya, Africa with the Leakey Foundation.

Epilogue

During the past four and one-half decades Roberts has been part of the explosive growth of physical organic chemistry. The important conceptual influences that shaped this field created one of the great advances in the history of science. The study of reactive intermediates as a method of generalizing organic reactions and the importance of characterizing their kinetic behavior was developed

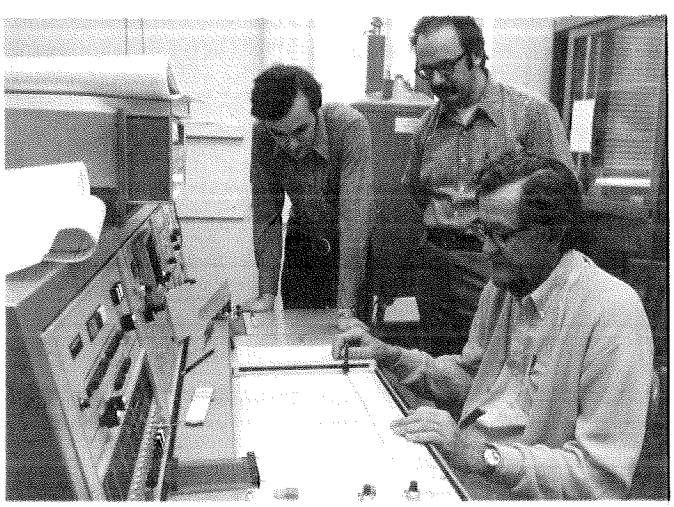

Fig. 11 - The Bruker WH-180 was a fantastic machine. Roberts' group enjoyed the luxury of a dedicated ^{15}N spectrometer which allowed them to run spectra as long as 72 hours.

by pioneers such as Roberts, his students and his contemporary colleagues — in both industry and academia — researchers of extraordinary intellect, individualism, discipline and commitment. The mechanistic framework that developed during those decades has profoundly influenced the way we currently *think and teach* organic chemistry. This lasting legacy has changed the intellectual framework of the field.

Roberts' direct scientific contributions are significant and for these he has been greatly honored by his profession.[18] Likely, this pioneering research (isotope-position rearrangements, benzyne, nonclassical cations, conformational analysis by NMR) will lose its identity and merge with the great body of knowledge in organic chemistry. My own feeling is that Roberts' contribution to science is much larger than that. He has shown by example that, in addition to research output (460 scientific papers published), teaching

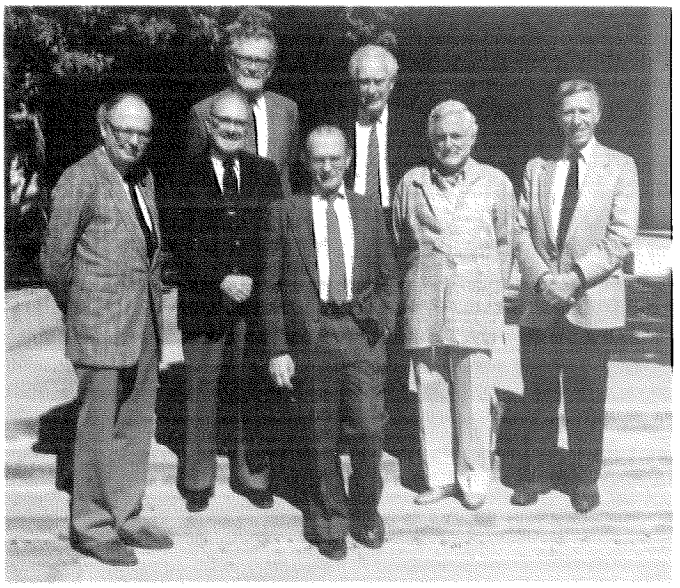

Fig. 12 - Great men of science who have contributed so much to the field of organic chemistry pose at a symposium to honor Professor William S. Johnson at Stanford University in 1986. (Left to right) Albert Eschenmoser, Bill Johnson, Jack Roberts, Gilbert Stork, Sir Derek Barton, Carl Djerassi, Conrad Block.

students, writing textbooks,[19] serving on editorial boards[20] and national committees,[21] consulting for industry, administering departments and universities are not only important, they form the mainstream of scientific life. The human element is clearly very important to John D. Roberts. This balance of achievement is a model for all young scientists.

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18) Professor Roberts has been awarded honorary degrees from the University of Munich and Temple University. He is a member of the American Chemical Society, the American Academy of Arts and Sciences, the American Philosophical Society, and the National Academy of Sciences. He received the ACS Award in Pure Chemistry (1954), the Harrison Howe Award (1957), the 1957 UCLA Alumni Professional Achievement Award in the Field of Science, the Roger Adams Medal and Award in Organic Chemistry (1967), the Nichols Award (1972), the Richard C. Tolman Medal (1975), the Michelson-Morely Award (1972), the James Flack Norris Award (1979), the Linus Pauling Award (1980), the Theodore William Richards Medal (1982), the Willard Gibbs Gold Medal (1983), the American Academy of Achievement Golden Plate Award (1984), and the Priestley Medal (1987).
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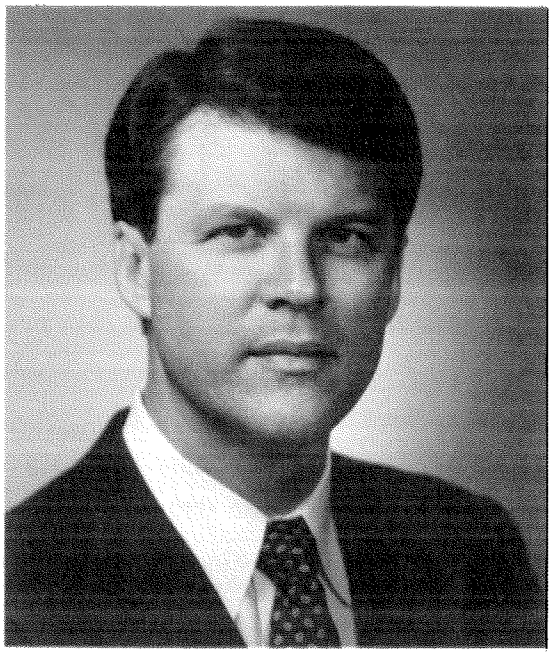
20) Professor Roberts has been a member of the editorial board of the American Chemical Society. He has been Editorial Consultant of W.A. Benjamin, Inc., of which he also served as Director, from the founding of the company until the spring of 1967. He has been a Director of Organic Synthesis, Inc. since 1977, and presently serves on the editorial boards of *Tetrahedron, Nouveau Journal de Chimie, Magnetic Resonance in Chemistry* and *Spectroscopy.* Since 1984, Professor Roberts has been a member of the Council of the International Society of Magnetic Resonance, the Board of Directors of the Huntington Medical Research Institute, the Board of Directors of the Caltech Y, the Board of Trustees of the L.B.S. Leakey Foundation, and the Executive Committee of the American Academy of Arts and Sciences Western Section.
21) He was a member and chairman of both the Advisory Panel for Chemistry of the National Science Foundation (1957-1960) and the Advisory Committee of Mathematical and Physical Sciences of the NSF (1962-1966); he was Chairman of the National Academy of Sciences Section of Chemistry (1968-1971), Chairman of Class I (Mathematical and Physical Sciences) (1976-1980), and Member of the Council (1980-1983). He also served on the National Academy of Sciences Committee for the Survey of Chemistry, which produced the report "Chemistry: Opportunity and Needs", on the Panel on Scientific Communication and National Security, and on the Committee on Science, Engineering, and Public Policy. He was elected Councillor, Class I, of the American Philosophical Society in 1983.

About the Author



Professor Peter B. Dervan was trained as a physical organic chemist working with Jerome A. Berson at Yale University (1972). After postdoctoral research at Stanford, he joined the chemistry faculty at the California Institute of Technology in 1973 where he is now Bren Professor of Chemistry. Professor Dervan's research interests span physical organic and bioorganic chemistry. He is a member of the National Academy of Sciences and a Fellow of the American Academy of Arts and Sciences.

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17,610-9 Mercuric acetate, 98 + %, A.C.S. reagent 100g $19.45; 500g $75.25; 6 x 500g $382.75
19,734-3 Methyllithium, 1.4*M* in diethyl ether 100ml $10.25; 800ml $43.65; 8l $337.25; 18l $631.25
P2,238-9 Phenyl dichlorophosphate 25g $8.65; 100g $24.90
20,967-8 Phosphorus, powder, red, 99% 100g $10.35; 500g $17.85
24,035-4 Propionic acid, 99 + %, GOLD LABEL 100g $8.85
24,031-1 Propionic anhydride, 99 + % 50g $10.65
P5,155-9 Propionyl chloride, 97% 500g $19.40
T2,250-0 *N,N,N′,N′*-Tetramethylethylenediamine, 99 + % (TMEDA) 100g $9.25; 500g $30.55
24,024-9 Thiophenol, 99 + % 50g $29.65
23,478-8 Tributyltin hydride, 97% 10g $12.25; 50g $45.25
13,206-3 Triethylamine, 99% 500g $13.90; 2kg $30.55
19,998-2 Tris(triphenylphosphine)rhodium(I) chloride (Wilkinson's catalyst) 1g $25.15; 5g $93.25

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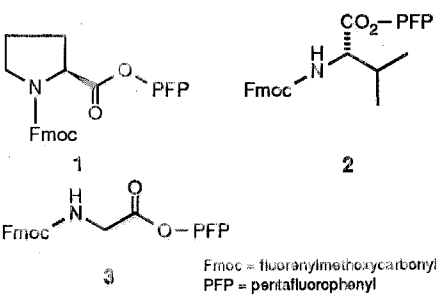
NEW *Fmoc/PFP Amino Acids for Peptide Synthesis*



Fmoc-protected amino acid activated esters for both solid-[1,2] and solution-phase peptide synthesis.[3]

(1) Atherton, E.; Sheppard, R.C. *Chem. Commun.* **1985,** 165. (2) Kisfaludy, L.; Schön, I. *Synthesis* **1983,** 325. (3) *Idem ibid.* **1986,** 303.

33,242-9 *N*-(9-Fluorenylmethoxycarbonyl)-L-proline pentafluorophenyl ester, 97% (1) 1g $18.00
33,243-7 *N*-(9-Fluorenylmethoxycarbonyl)-L-valine pentafluorophenyl ester, 97% (2) 1g $18.00; 5g $60.00
33,529-0 *N*-(9-Fluorenylmethoxycarbonyl)glycine pentafluorophenyl ester, 97% (3) 1g $18.00

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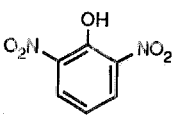

D19,880-3 2,6-Dinitrophenol, 97% 1g $10.00; 5g $35.00

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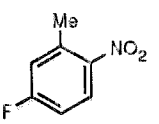

F1,240-5 5-Fluoro-2-nitrotoluene, 97% 10g $12.00; 50g $50.00